

IN THE UNITED STATES PATENT AND TRADEMARK OFFICE

In re Patent Application of:

Confirmation No.: 9742

Jörg Kowalczyk, et al.

Group Art Unit: 1623

Serial No.: 10/555,714

Examiner: Layla D. Bland

Filed: July 27, 2006

For: METHOD FOR SELECTIVE CARBOHYDRATE OXIDATION USING
SUPPORTED GOLD CATALYSTS

VIA EFS-WEB

Commissioner for Patents

P.O. Box 1450

Alexandria, Virginia 22313-1450

THIRD DECLARATION UNDER 37 C.F.R. § 1.132 OF DR. ALI REZA HAJI BEGLI

Sir:

1. I am the same individual who previously executed a first declaration on 23 June 2009 that, I am informed, was submitted by our U.S. patent counsel to the U.S. Examiner in charge of this application together with a “Response to Final Office Action” on 25 June 2009. I additionally executed a second declaration (on 11 June 2010) that, I am also informed, was submitted by our U.S. counsel together with an “Amendment Under 37 C.F.R. §1.114” on 16 June 2010. My education and employment experience are as set forth in my first declaration. As, moreover, indicated therein, I am a co-inventor of the present application, employed by Südzucker Aktiengesellschaft Mannheim/Ochsenfurt, the owner by assignment of the application. I am making this declaration in support of the patentability of the claims presently pending in the application.

2. I have read and I am thus familiar with the non- final Office Action issued by the Examiner having a mailing date of 24 August 2010, as well the references cited therein, in combination, to reject the present claims. Furthermore, I have additionally reviewed and considered the matters discussed in the Examiner’s 3 January 2011 Interview Summary of the telephone interview held by our U.S. patent counsel with the Examiner on 21 December 2010.

3. I understand from our U.S. counsel that during the telephone interview the Examiner agreed that the evidence previously provided in this case does demonstrate an

improvement on the durability of the metal oxide supported gold catalyst compared to the carbon supported gold catalyst according to Biella (Journal of Catalysis, 2002), however the showing is not sufficiently broad to support the patentability of claims having the scope of those presently pending in the application. In particular I understand that the Examiner's position is that the unexpected technical effect, namely the higher durability of the claimed metal oxide supported gold catalysts, has not yet been proven for a sufficient variety of different metal oxide supports, at different pH values and when different carbohydrates are oxidized selectively at the C1-atom.

Therefore, I submit hereby further experimental data, obtained by me or under my direction and control, that I submit effectively demonstrates that the improved durability of the catalysts according to the present invention in comparison to the carbon supported gold catalysts according to Biella et al. over the full scope of claims presently pending in this application.

4. Outline of Experiments Demonstrating the Greater Durability of the Claimed Catalysts as Compared to Catalysts Disclosed in the Closest Prior Art (Biella et al.)

In a first approach, glucose was oxidized using a 1% Au/Al₂O₃ catalyst at an adjusted pH measuring: (i) 7, (ii) 8 or (iii) 9.5, in 10 consecutive batches for each group. The reaction time until the completion of the oxidation was used as the measure of the catalytic activity.

In a second approach, glucose was oxidized with use of a 1% Au catalyst on different metal oxide-supports at different pH's in 4 consecutive batches.

In a third approach, different carbohydrates were oxidized by 1% Au catalyst having different metal oxide-supports at different pH over 4 consecutive batches.

4.1 Material and Methods

4.1.1 Oxidation procedure

To carry out carbohydrate oxidation 2.2 g of 1% Au/metal oxide catalyst prepared according to the invention was suspended in 500 ml of aqueous carbohydrate solution, thus corresponding to a ratio of carbohydrate to gold of 1,000. Oxygen was bubbled through the slurry at a rate of 20 ml/min (atmospheric pressure). The temperature was kept constant at 50°C (323 K) over the entire reaction. The pH was constantly maintained at the predefined value by adding dropwise NaOH (0.3 mol/L). After completion of each batch the catalyst was washed and prepared for re-use.

Solutions with either (i) 4% by weight glucose, (ii) 8% by weight of lactose or (iii) 8% by weight of a maltose rich syrup comprising 5 weight % maltotriose and 10 % oligosaccharides were used as aqueous carbohydrate solutions (each 500 ml per batch).

As the metal oxide, support MgO (2nd principal group), Al₂O₃ (3rd principal group), SiO₂ (4th principal group), TiO₂ (4th subgroup) and CeO₂ (lanthanoids) were employed alternatively.

As stated above, the ratio of carbohydrate or mixture of carbohydrate to gold was adjusted to 1,000 to be in conformity with the ratio used in the closest prior art of Biella.

4.1.2 The 100% conversion of carbohydrate to C1-oxidized carbohydrate was achieved when no further NaOH had to be added to maintain the adjusted pH value. The reaction time was measured (in minutes) and is set forth in table 1 attached hereto.

4.2 The reaction time so measured correlates with the catalysts' activity and durability. With a catalyst having a high activity the time to convert a given amount of glucose to gluconic acid will be short. A high durability of a catalyst is characterized by a substantially constant, particularly not increasing, reaction time or activity over repeated batches.

4.3 It is noted that the reaction times referring to the catalysts according to Biella et al. (1 % Au/C) were taken from the figures 9a to 9c of the subject reference. These reaction times are illustrated in figure 1 provided herewith.

4.4 Results

The results of the experiments are set forth in figures 2 to 6 and in table 1:

Figure 2 illustrates the reaction time of 1% gold catalysts with different metal oxide supports according to the invention, namely Al₂O₃, TiO₂, CeO₂, MgO and SiO₂, at a pH of 9.5 over four batches.

Figure 3 shows the reaction time of 1% Au/Al₂O₃ catalyst at different pH values over at least four batches, when different carbohydrates are oxidized.

Figure 4 shows the reaction time of 1% Au/TiO₂ catalyst at different pH values over four batches, when different carbohydrates are oxidized.

Figure 5 shows the reaction time of 1% Au/CeO₂ catalyst at different pH values over four batches, when different carbohydrates are oxidized.

Also as shown in Figures 3 to 5, the oxidation of lactose and maltose rich syrup was not performed at pH 8 due to the short period of time. However, for simplifying the comparison of the figures placeholders are used.

Figure 6 and table 1 show an overview of the results obtained. In figure 6 the black lines represent the reaction times of the catalysts according to Biella (diamond for pH = 7;

square for pH = 8; triangle for pH = 9.5). The white lines show the reaction times of the catalysts according to the present invention (diamond for pH = 7; square for pH = 8; triangle for pH = 9.5).

4.5 Discussion

The activity of the catalysts according to the invention is almost constant over the repeated batches at different pH. In contrast thereto, the catalysts according to Biella et al. show a remarkable decrease in activity already after three or four batches. The higher durability of the catalysts according to the invention compared to those disclosed in Biella et al. is also present when different carbohydrates were oxidized.

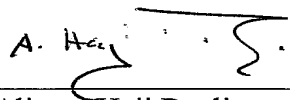
4.6 Summary

The substantial differences demonstrated in the attachments to this declaration clearly demonstrate to a skilled artisan in this field that the catalysts according to the presently pending claims exhibit an unexpected greater durability than those produced according to the teachings of Biella et al.

5. I further declare all statements made herein of my own knowledge are true and that all statements made on information and belief are believed to be true, and further, these statements were made with the knowledge that false statements and the like so made are punishable by fine or imprisonment, or both, under section 1001 of Title 18 of the United States Code, and that such willful false statements made herewith may jeopardize the validity of the application or any patent issuing thereon.

Date: 18.02.2011

By:


Dr. Alireza Haji Begli

carbon support (Biella)

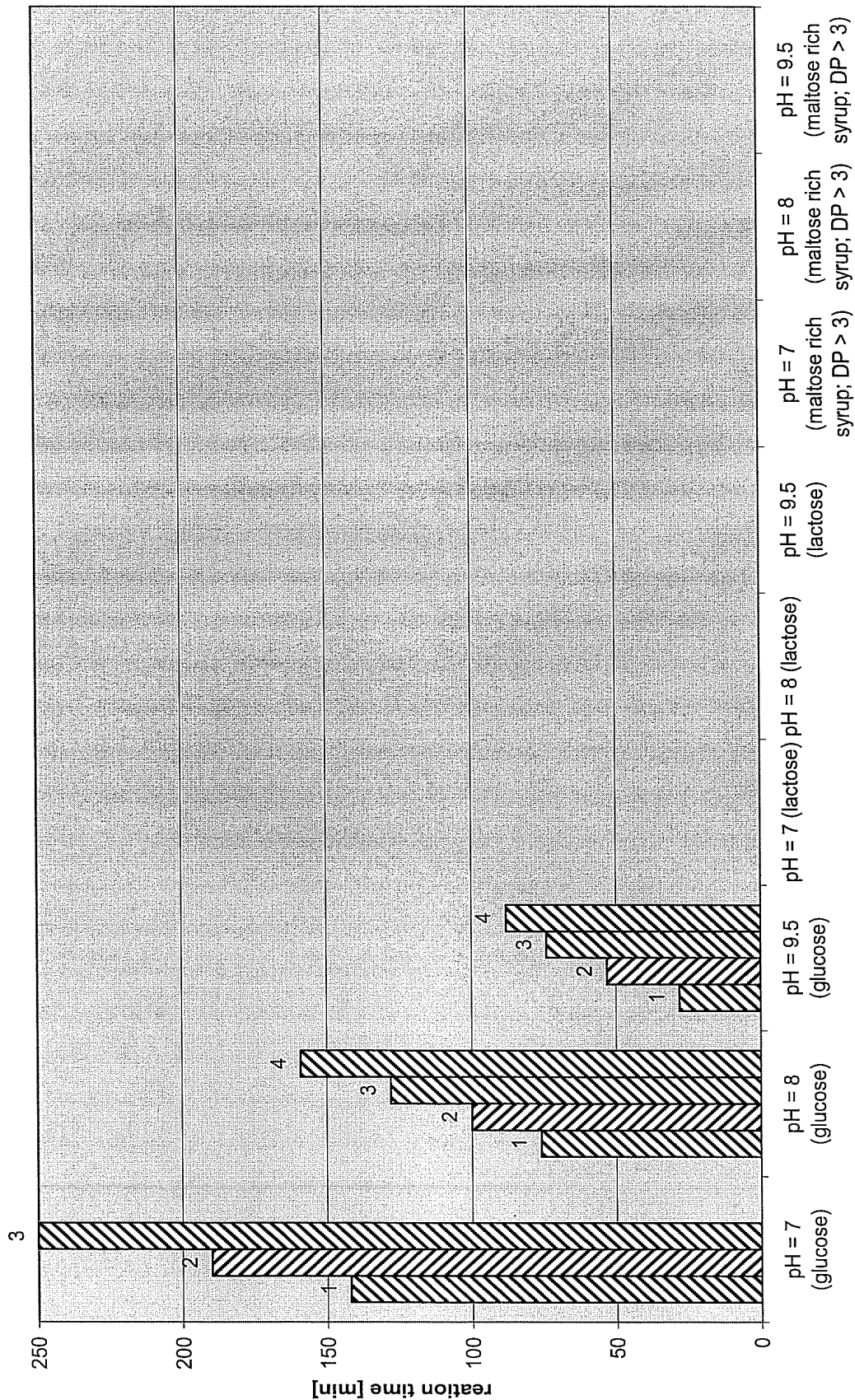


Figure 1

metal oxide support (invention) at pH = 9.5

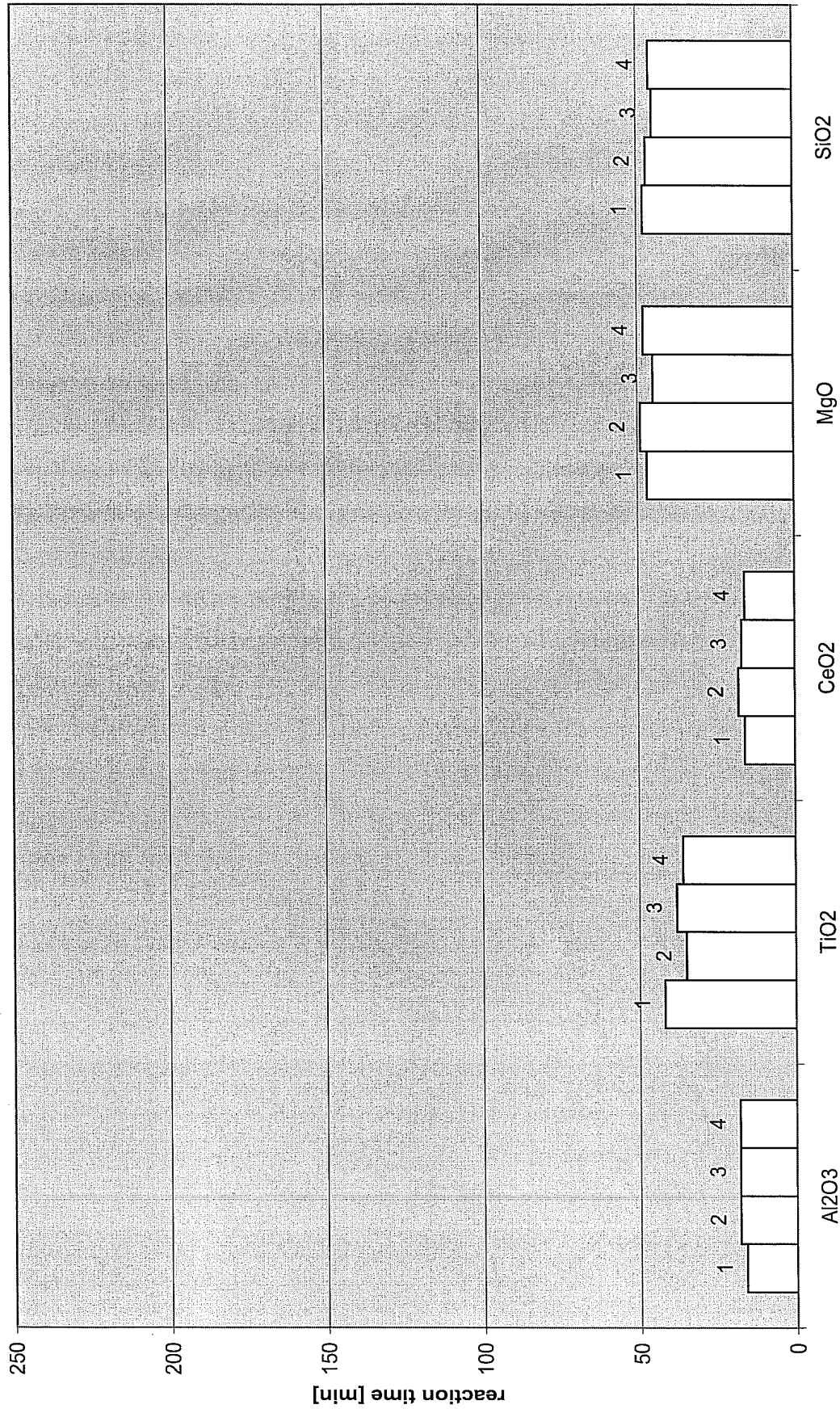


Figure 2

1% Au/Al₂O₃ (invention)

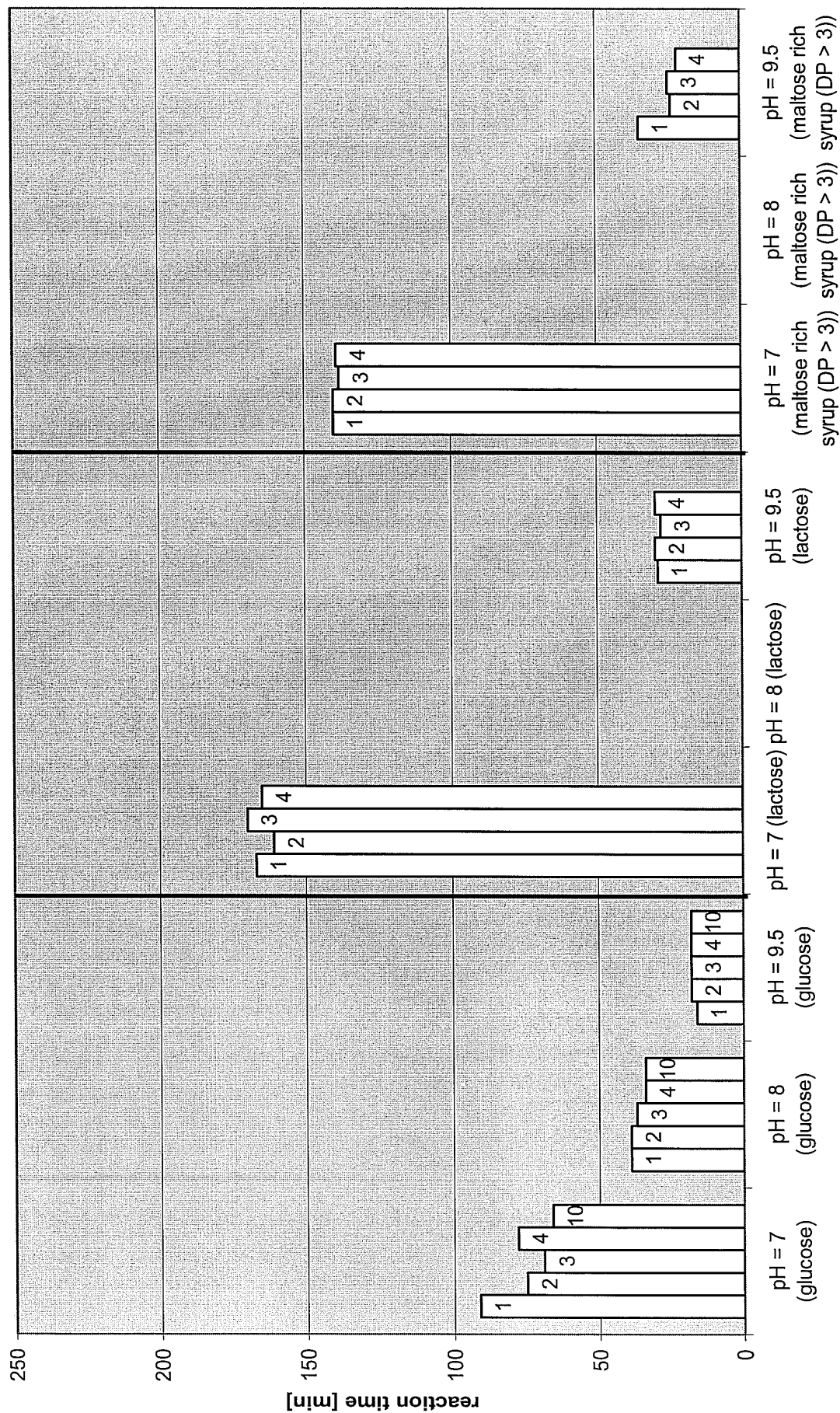


Figure 3

1% Au/TiO₂ (invention)

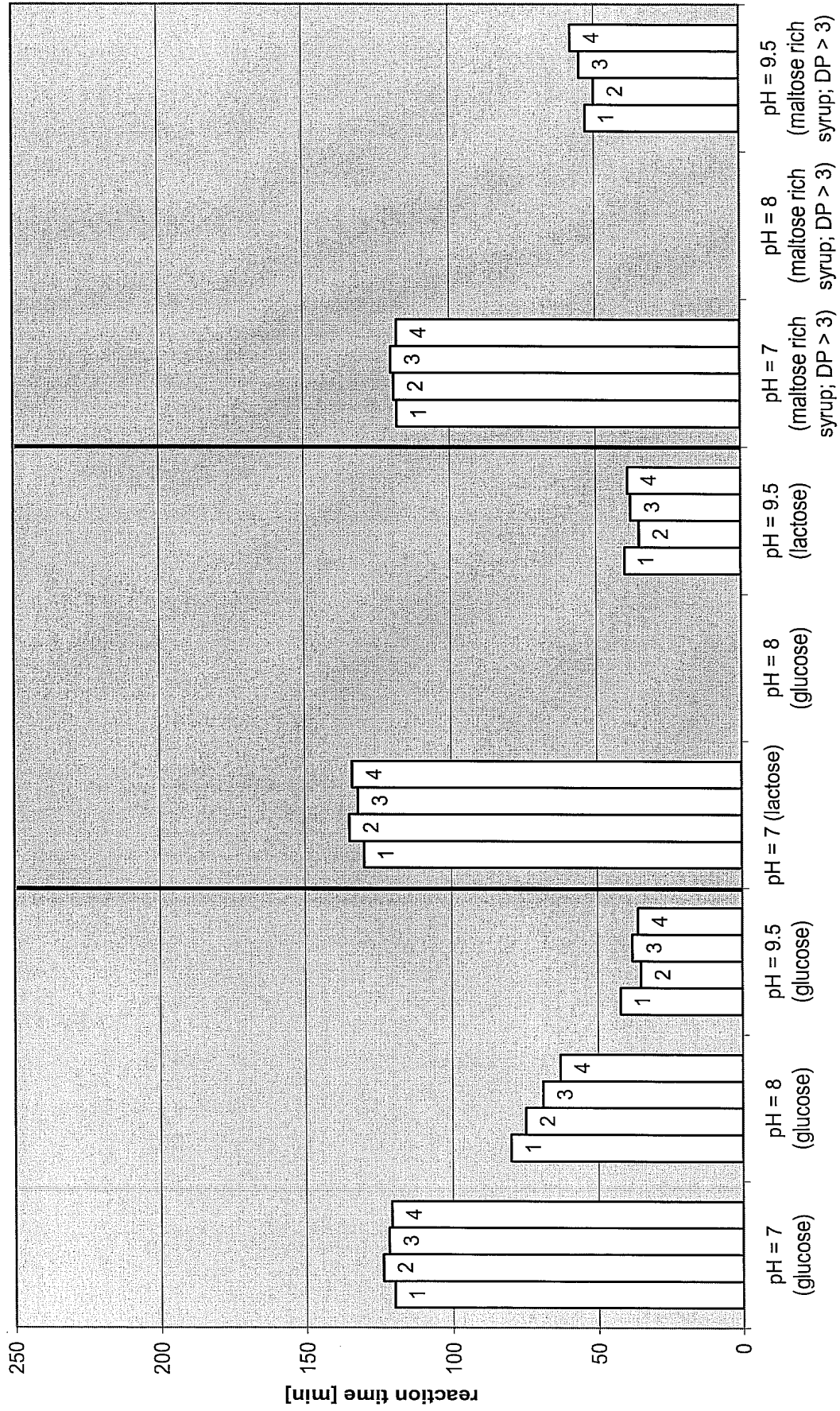


Figure 4

1%**AuCeO2** (invention)

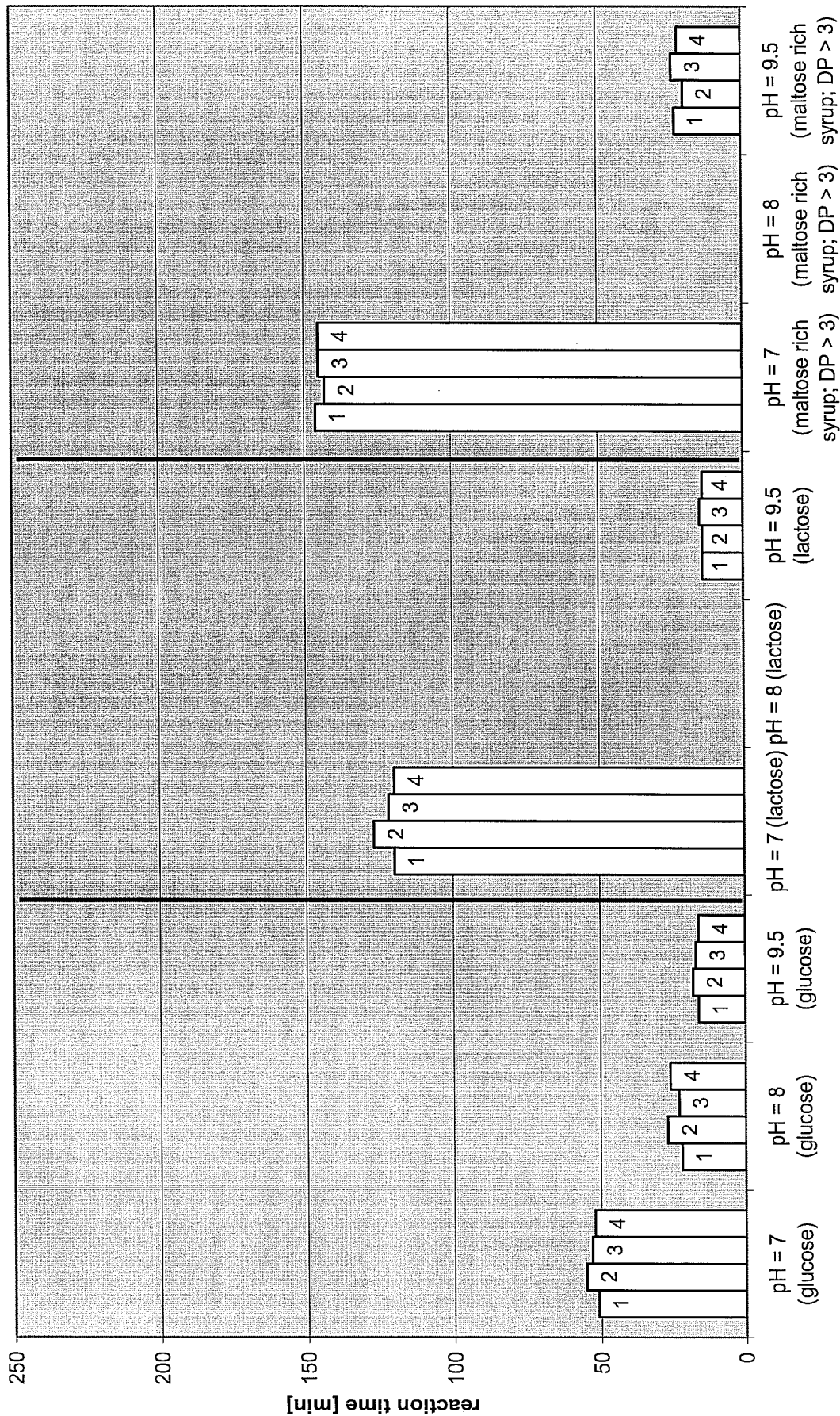


Figure 5

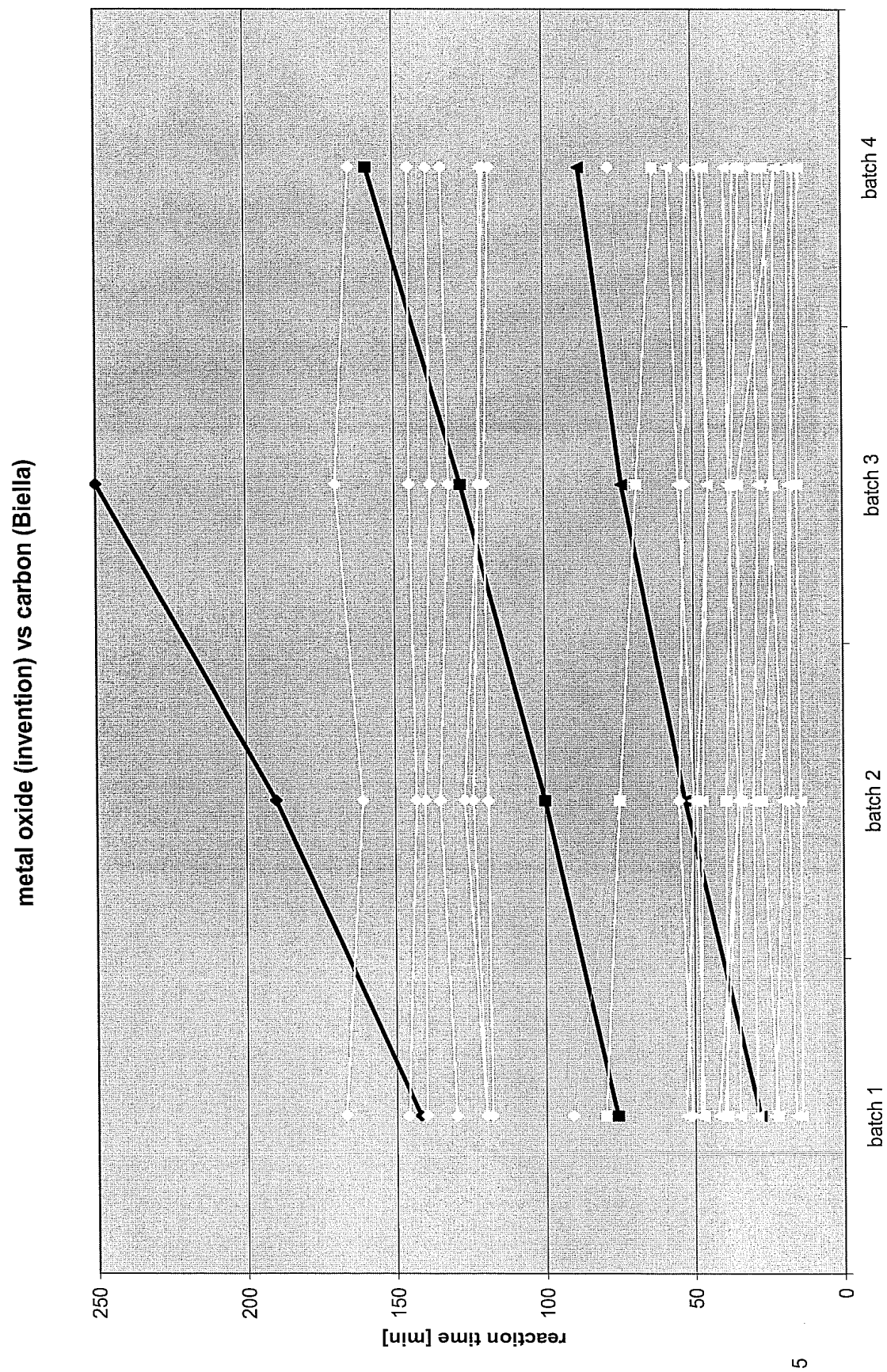


Figure 6

	batch 1	batch 2	batch 3	batch 4	batch 10
1% Au/C; pH = 7; glucose	142	190	250		
1% Au/C; pH = 8; glucose	76	100	128	159	
1% Au/C; pH = 9.5; glucose	28	53	74	88	
1% Au/Al ₂ O ₃ ; pH = 7; glucose	91	75	69	78	66
1% Au/Al ₂ O ₃ ; pH = 8; glucose	39	39	37	34	34
1% Au/Al ₂ O ₃ ; pH = 9.5; glucose	16	18	18	18	18
1% Au/Al ₂ O ₃ ; pH = 7; lactose	167	161	170	165	
1% Au/Al ₂ O ₃ ; pH = 9.5; lactose	29	30	28	30	
1% Au/Al ₂ O ₃ ; pH = 7; maltose rich syrup	140	140	138	139	
1% Au/Al ₂ O ₃ ; pH = 9.5; maltose rich syrup	35	24	25	22	
1% Au/TiO ₂ ; pH = 7; glucose	120	124	122	121	
1% Au/TiO ₂ ; pH = 8; glucose	80	75	69	63	
1% Au/TiO ₂ ; pH = 9.5; glucose	42	35	38	36	
1% Au/TiO ₂ ; pH = 7; lactose	130	135	132	134	
1% Au/TiO ₂ ; pH = 9.5; lactose	40	35	38	39	
1% Au/TiO ₂ ; pH = 7; maltose rich syrup	118	119	120	118	
1% Au/TiO ₂ ; pH = 9.5; maltose rich syrup	53	50	55	58	
1% Au/CeO ₂ ; pH = 7; glucose	51	55	53	52	
1% Au/CeO ₂ ; pH = 8; glucose	22	27	23	26	
1% Au/CeO ₂ ; pH = 9.5; glucose	16	18	17	16	
1% Au/CeO ₂ ; pH = 7; lactose	120	127	122	120	
1% Au/CeO ₂ ; pH = 9.5; lactose	14	14	15	14	
1% Au/CeO ₂ ; pH = 7; maltose rich syrup	146	143	145	145	
1% Au/CeO ₂ ; pH = 9.5; maltose rich syrup	23	20	24	22	
1% Au/MgO; pH = 9.5; glucose	47	49	45	48	
1% Au/SiO ₂ ; pH = 9.5; glucose	48	47	45	46	

Table 1